

Thermomechanical effects in uniformly aligned dye-doped nematic liquid crystals

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(Dated: February 2, 2008)

Abstract

We show theoretically that thermomechanical effects in dye-doped nematic liquid crystals when illuminated by laser beams, can become important and lead to molecular reorientation at intensities substantially lower than that needed for optical Fréedericksz transition. We propose a 1D model that assumes homogenous intensity distribution in the plane of the layer and is capable to describe such a thermally induced threshold lowering. We consider a particular geometry, with a linearly polarized light incident perpendicularly on a layer of homeotropically aligned dye-doped nematics.

PACS numbers: 05.45.-a, 42.70.Df, 42.65.Sf

I. INTRODUCTION

Optically induced temperature changes in liquid crystals are at the origin of interesting nonlinear behaviors [1]. Indeed, due to the light absorption, thermal effects can change the physical properties of the liquid crystal, which in turn affects the light propagation in the medium. Among the examples of thermal effects, we can distinguish between a direct change of the refractive indices, often referred to as thermal indexing, and the variation with the temperature of other physical parameters, such as the elastic constants, which may influence the light propagation as well. In the latter case, thermal effects might be responsible for director reorientation, and are thus referred to as thermomechanical effects.

Previously, light-induced thermomechanical effects have been largely investigated in cholesteric liquid crystals, where these effects were originally related with the absence of the right-left symmetry [2]. Then it became clear that such effects might also exist in systems which possesses this symmetry, such as the nematic liquid crystals (NLC). As was shown in [3, 4], thermomechanical effects do give a contribution to the director, heat and Navier-Stokes equations. These thermomechanical terms are given by nonlinear combinations with respect to temperature, director and velocity gradients and represent nonlinear cross couplings between them. The first question which arose is to suggest an experiment for measuring the magnitude of thermomechanical coefficients. Indeed, it is hardly possible to find a setup which leads to the contribution of one of the coefficients only. Since for uniformly aligned nematics no thermomechanical effects are expected, the hybrid-oriented nematics were used in the experiments [5, 6]. There, the hydrodynamic flow appeared as a result of the applied temperature gradient, which allowed to find the magnitude for the thermomechanical coefficients. Laser induced thermomechanical effects in dye-doped nematics have been envisaged in preliminary experiments [7]. However, it was not possible to derive a definitive conclusion due to the main difficulty of separating thermomechanical contributions from the light-induced molecular torque. Another problem when performing these experiments is that of avoiding to approach the nematic-isotropic transition, where an enhancement of the nonlinear optical response of dye-doped nematics could take place because of other effects, such as the weakening of the anchoring [8].

Here, we study theoretically the thermomechanical effects which occur in uniformly aligned dye-doped nematic liquid crystals. This happens when the sample is illuminated

by a laser beam with a wavelength in the absorption band of the dye, which causes a significant heating of the liquid crystal layer. Note that the absorbtion is negligible for pure nematics, so that thermomechanical effects are significant only for dye-doped nematics. Indeed, in the presence of dye-doping thermal heating leads to an additional torque which acts onto the director together with the light-induced torque. This additional torque will help either to destabilize or stabilize the initial orientation which is determined by the ratio of the thermomechanical coefficients. We show that the additional torque might be strong enough to induce the molecular reorientation well before the onset of the light-induced molecular reorientation, the so called optical Fréedericksz transition (OFT) for pure liquid crystals [9] and Janossy effect for dye-doped liquid crystals [10, 11]. We account for the possible decrease of the reorientation threshold by a 1D model that assumes homogenous intensity distribution in the plane of the layer (the plane wave approximation) and includes the light absorption into the hydrodynamic equations for the nematics.

II. THEORETICAL MODEL

In dye-doped nematics, contrary to the pure ones, the OFT lowering might happen not only because of the well known Janossy effect [10, 11] but also because thermomechanical effects may add a significant contribution. Indeed, as a consequence of light absorption, light propagation in dye-doped nematics causes significant heating of the LC. When the intensity of a beam is sufficiently large and temperature gradient becomes nonzero, an additional force acting on the director and additional terms in the stress tensor in Navier-Stokes equation for the velocity appear due to thermomechanical effect [3, 4]. The equation for the velocity is coupled with the director equation, so any dynamical process that leads to director reorientation will also induce flow even in the absence of pressure gradients. It should be noted that the terms which describe the thermomechanical effects appear not only in the director but also in Navier-Stokes equations. Thus, neglecting the velocity equations and considering only the director equation may lead to misleading results, even though this simplification has been done for many studies in the context of light induced instabilities.

In our case, we considered the full problem with the velocity equations coupled to the director reorientation equation. However, in order to calculate the shift of the OFT threshold due to the thermomechanical effect we employed another frequently used simplification,

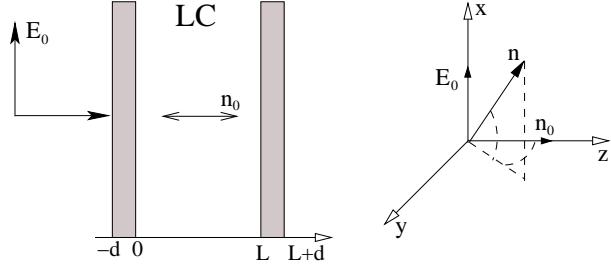


FIG. 1: Geometry of the setup: a linearly polarized light along the **x**-direction is at a normal incidence on a nematic LC layer; $\mathbf{n}_0 \parallel \mathbf{z}$ is the director of the initial unperturbed alignment (homeotropic state).

namely that all variables depend only on one coordinate, which is transversal to the plane of the nematic layer (1D assumption). We thus considered a linearly-polarized plane wave incident perpendicularly on a layer of a dye-doped nematic of thickness L that is sandwiched between two substrates of thickness d (see Fig. 1). The cell has initially homeotropic alignment (with strong homeotropic anchoring at the boundaries) and is placed in a thermostage with a temperature T_0 on both sides. The light is polarized along the **x**-direction and propagates along the positive **z**-axis.

A. Heat equation

For simplicity we also assumed that i) the attenuation of the light inside the nematic is small [i.e. $I(z) \simeq I_0$, where I_0 is the incident intensity]; ii) the transversal heat flow occurring in the (\mathbf{x}, \mathbf{y}) plane is neglected, i.e. the temperature profile depends only on z , $T = T(z)$. Since we deal with the plane-wave approximation the following 1D steady-state heat conductivity equations in the nematic and substrates can be written (note that an absorption takes place only inside the nematic):

$$\begin{aligned} \kappa_s \partial_z^2 T &= 0, \quad -d \leq z \leq 0 \text{ or } L \leq z \leq L + d \\ \kappa_{\parallel} \partial_z^2 T &= -\alpha_{\perp} I_0, \quad 0 \leq z \leq L, \end{aligned} \quad (1)$$

where κ_{\parallel} is the parallel component of the heat conductivity tensor of the nematic, κ_s is the heat conductivity of the substrates and α_{\perp} is the absorption coefficient for the ordinary light. We then write the boundary conditions given by continuity conditions of the temperature

and the heat flow at the substrate-nematic interfaces ($z = 0$ and $z = L$):

$$T_{s1}|_{z=0} = T_N|_{z=0}, \quad \kappa_s \partial_z T_{s1}|_{z=0} = \kappa_{||} \partial_z T_N|_{z=0} \quad (2)$$

$$T_N|_{z=L} = T_{s2}|_{z=L}, \quad \kappa_{||} \partial_z T_N|_{z=L} = \kappa_s \partial_z T_{s2}|_{z=L}. \quad (3)$$

In this simplest model the temperature profile is linear inside the substrates and has a parabolic form inside the nematic which is symmetric with respect to the center of the layer $z = L/2$. The maximal temperature is at the center of the layer and the maximal temperature difference inside the nematic is $\Delta T_{max} = \alpha_{\perp} I_0 L^2 / (8\kappa_{||})$. The temperature gradient inside the nematic can be written as

$$\partial_z T_N = \beta \rho (L - 2z), \quad \text{with } \beta = \frac{\alpha_{\perp} I_F}{2\kappa_{||}}, \quad (4)$$

where $\rho = I_0/I_F$ is the incident intensity normalized to the threshold intensity of the OFT for a dye-doped nematic [12]

$$I_F = \frac{\pi^2}{L^2} \frac{c(\varepsilon_{\perp} + \varepsilon_a)K_3}{\varepsilon_a \sqrt{\varepsilon_{\perp}} \eta}, \quad \eta = \frac{\varepsilon_a + \zeta}{\varepsilon_a}. \quad (5)$$

Here $\varepsilon_a = \varepsilon_{||} - \varepsilon_{\perp}$ is the dielectric anisotropy and ε_{\perp} ($\varepsilon_{||}$) is the dielectric permittivity (at optical frequency) perpendicular (parallel) to \mathbf{n} , ζ phenomenologically describes the effect of certain dye dopants ($\zeta = 0$ in a pure LC), K_3 is the bend elastic constant of the nematic and c is the velocity of light in the vacuum.

It should be noted that the obtained solution (4) for the temperature profile inside the nematics is much simpler than in reality. The more complicated solution for Gaussian incident beams has been derived in [13] from 2D heat equation which includes transversal dependence. It turned out that for large Gaussian beams i.e. when the spot size w is much larger than the thickness of the layer L , $w \gg L$, the maximum temperature rise, ΔT_{max} , becomes proportional to the spot size. Such a behavior is not predicted by the 1D model and is owing to the transversal heat flow occurring in the plane of the layer. Nevertheless, the realization of the plane wave approximation in the experiment is difficult but not impossible task. One of the way to proceed is to enlarge the Gaussian beam to a size which is much larger than the medium working area (which is much stronger condition then $w \gg L$), so that the intensity can be considered uniform in the central part. Another possibility is that of using the so-called flat-top beams [14]. Both techniques are quite easily accessible and allow to attain the threshold for OFT. Preliminary experiments are running at present in our laboratory and will be reported elsewhere.

B. Linearized Navier-Stokes equation

The Navier-Stokes equation for the velocity \mathbf{v} can be written as [15]

$$\rho_m (\partial_t + \mathbf{v} \cdot \nabla) v_i = -\nabla_j (p \delta_{ij} + \pi_{ij} + T_{ij}^{visc} + T_{ij}^{TM}), \quad (6)$$

where ρ_m and p are the density and the pressure of the LC, respectively. π_{ij} is the Ericksen stress tensor [15]. The viscous stress tensor T_{ij}^{visc} in Eq. (6) is written in terms of the six Leslie coefficients α_i [15] and the thermomechanical tensor T_{ij}^{TM} is introduced in [3, 4]. Then, the incompressibility condition (ρ_m is constant) $\nabla \cdot \mathbf{v} = 0$ and the no-slip boundary conditions $\mathbf{v}|_{z=0,L} = 0$ immediately ensure that the z component of the velocity vanishes $\mathbf{v} = (v_x(z, t), v_y(z, t), 0)$, so \mathbf{v} is parallel to the plane of the layer. Moreover, all convective derivatives $\mathbf{v} \cdot \nabla$ vanish. The Navier-Stokes equation has been simplified then due to the fact that the director relaxation time

$$\tau = \frac{\gamma_1 L^2}{\pi^2 K_3} \quad (7)$$

differs by many orders of magnitude with the momentum diffusion time $\tau_{visc} = \rho_m L^2 / \gamma_1$, where $\gamma_1 = \alpha_3 - \alpha_2$ is the rotational viscosity. Typically $\tau_{visc} \sim 10^{-6} s$ and $\tau \sim 1 s$, so the slow variable of the system is the evolution of the director which enslaves the flow motion and, thus, the inertial terms in the Eq. (6) can be neglected. Taking into account that the light is polarized in the \mathbf{x} -direction, we need an equation for the x component only. The linearization of this equation around the homeotropic state ($n_x = 0, v_x = 0$) yields

$$\begin{aligned} & (\alpha_5 - \alpha_2 + \alpha_4) \frac{\partial_z v_x}{2} + \alpha_2 \partial_t n_x - \\ & a_{10} \partial_z n_x \partial_z T_N = C(t), \end{aligned} \quad (8)$$

where $C(t)$ is a function that does not depend on z and will be fixed by the boundary conditions. Here the first two terms on the left-hand side of Eq. (8) come from the viscous stress tensor whereas the last one is the contribution from the thermomechanical stress tensor with the thermomechanical coefficient a_{10} . (Note that a_{10} in [4] is related to the ξ_i from [3] as $a_{10} = (\xi_8 - \xi_4)/4$.) The unknown function $C(t)$ can be determined by integrating Eq. (8) across the layer. Finally, the velocity gradient $\partial_z v_x$ can be expressed in terms of the

director and the temperature gradient [which is given by Eq. (4)] as:

$$\partial_z v_x = \frac{2}{\alpha_5 - \alpha_2 + \alpha_4} \times \quad (9)$$

$$[C(t) - \alpha_2 \partial_t n_x + a_{10} \beta \rho (L - 2z) \partial_z n_x] \\ C(t) = \frac{1}{L} \left(\alpha_2 \int_0^L \partial_t n_x dz - 2a_{10} \beta \rho \int_0^L n_x dz \right). \quad (10)$$

C. Linearized director equation. Adiabatic elimination of the flow field

The equation for the director \mathbf{n} is

$$\gamma_1 (\partial_t + \mathbf{v} \cdot \nabla - \boldsymbol{\omega} \times) \mathbf{n} = -\underline{\delta}^\perp (\gamma_2 \underline{\underline{A}} \mathbf{n} + \mathbf{h} - \mathbf{g}^{TM}), \quad (11)$$

where $\gamma_2 = \alpha_3 + \alpha_2$. \mathbf{h} is the molecular field obtained from the variational derivatives of the free energy density F , which consists of the elastic and the electrical parts [15]. The projection operator $\underline{\delta}^\perp = \delta_{ij} - n_i n_j$ in Eq. (11) ensures conservation of the normalization $\mathbf{n}^2 = 1$. Here A_{ij} is the symmetric strain-rate tensor and the vector \mathbf{N} gives the rate of change of the director relative to the fluid. In Eq. (11) \mathbf{g}^{TM} is the contribution of the thermomechanical effect to the force acting on the director [3].

We then linearized Eq. (11) around the homeotropic state and obtained the following equation for n_x

$$\gamma_1 \partial_t n_x + \alpha_2 \partial_z v_x = \quad (12) \\ K_3 \left[\partial_z^2 n_x + \left(\frac{\pi}{L} \right)^2 \rho n_x \right] - \frac{\xi_4}{2} \partial_z n_x \cdot \partial_z T_N.$$

Note that the first term in the square brackets on the right-hand side of Eq. (12) stems from the linearization of \mathbf{h} whereas the last one after the linearization of \mathbf{g}^{TM} .

From here on we will use normalized time $t \rightarrow t/\tau$ [where τ is the director relaxation time, see Eq. (7)], length $z \rightarrow \pi z/L$ (the same symbols will be kept). We will also introduce dimensionless viscosity coefficients $\alpha'_i = \alpha_i/\gamma_1$. Eliminating the velocity gradient from Eq. (12) with the help of Eq. (9) and using the expression for the temperature gradient (4), the following equation for n_x can be derived

$$\partial_z^2 n_x + \rho n_x - (1 - b) \partial_t n_x - d_1 \rho (\pi - 2z) \partial_z n_x - \quad (13) \\ \frac{b}{\pi} \left\{ \int_0^\pi \partial_t n_x dz - \frac{2d_2 \rho}{\alpha'_2} \int_0^\pi n_x dz \right\} = 0,$$

where b , d_1 and d_2 are dimensionless parameters defined as follows:

$$b = \frac{2\alpha_2'^2}{\alpha_5' - \alpha_2' + \alpha_4'}, \quad d_1 = \delta_1 \psi, \quad d_2 = \delta_2 \psi. \quad (14)$$

Here ψ depends on the absorption and $\delta_{1,2}$ are the algebraic combinations of the thermomechanical coefficients given by

$$\begin{aligned} \psi &= \frac{\beta\tau}{\gamma_1} = \frac{\alpha_\perp c(\varepsilon_\perp + \varepsilon_a)}{2\kappa_{||}\varepsilon_a\sqrt{\varepsilon_\perp}\eta}, \\ \delta_1 &= \left(\frac{b a_{10}}{\alpha_2'} + \frac{\xi_4}{2} \right), \quad \delta_2 = a_{10}. \end{aligned} \quad (15)$$

D. Linear stability analysis of the homeotropic state

We look for solutions of Eq. (13) of the form

$$n_x(z, t) = n_x(z)e^{\sigma t}, \quad (16)$$

where σ is the growth rate and obtain from Eqs. (13)

$$\begin{aligned} \partial_z^2 n_x + [\rho - \sigma(1 - b)] n_x - d_1 \rho (\pi - 2z) \partial_z n_x - \\ \frac{b}{\pi} \left(\sigma - \frac{2d_2 \rho}{\alpha_2'} \right) \int_0^\pi n_x dz = 0. \end{aligned} \quad (17)$$

It should be noted that Eq. (17) reduces to the classical linearized equation for the OFT when the flow and the thermomechanical effect are neglected by putting $b = 0$ and $d_1 = d_2 = 0$.

Taking into account the boundary conditions $n_x|_{z=0,\pi} = 0$, Eq. (17) is solved by

$$n_x = \frac{A}{\rho - \sigma(1 - b)} \left\{ -1 + e^{d_1 \rho (\pi - z) z} \frac{{}_1F_1 \left[\frac{1}{2} - \frac{\rho - \sigma(1 - b)}{4d_1 \rho}, \frac{1}{2}, \frac{d_1 \rho}{4} (\pi - 2z)^2 \right]}{{}_1F_1 \left[\frac{1}{2} - \frac{\rho - \sigma(1 - b)}{4d_1 \rho}, \frac{1}{2}, \frac{d_1 \rho \pi^2}{4} \right]} \right\}, \quad (18)$$

where ${}_1F_1$ is the confluent hypergeometric function [16] and A is some constant which depends on the director itself

$$A = -\frac{b}{\pi} \left(\sigma - \frac{2d_2 \rho}{\alpha_2'} \right) \int_0^\pi n_x dz. \quad (19)$$

Substituting the solution for n_x (18) into Eq. (19) the equation for the growth rate σ versus incident intensity ρ can be derived (note that A will be cancelled). Substituting $\sigma = 0$ into

this equation, the following transcendental equation for the critical intensity ρ_c has been obtained

$$\frac{\int_0^\pi e^{d_1 \rho_c (\pi - z) z} \cdot {}_1F_1 \left[\frac{1}{2} - \frac{1}{4d_1}, \frac{1}{2}, \frac{d_1 \rho_c}{4} (\pi - 2z)^2 \right] dz}{{}_1F_1 \left[\frac{1}{2} - \frac{1}{4d_1}, \frac{1}{2}, \frac{d_1 \rho_c \pi^2}{4} \right]} = \pi \left(1 + \frac{\alpha'_2}{2 b d_2} \right). \quad (20)$$

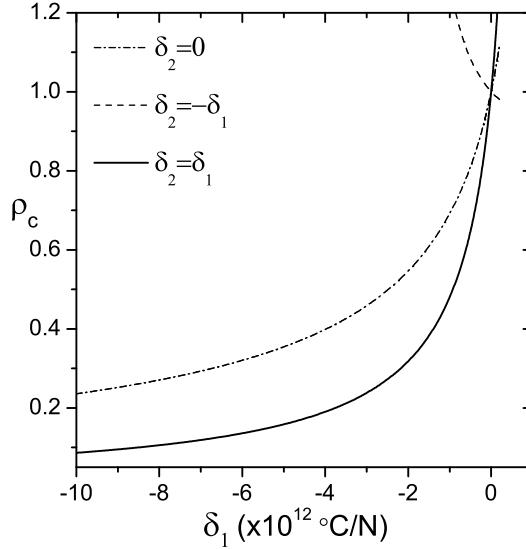


FIG. 2: The critical intensity ρ_c versus parameter δ_1 under assumptions that $\delta_2 = \delta_1$ (solid line), $\delta_2 = 0$ (dot-dashed line), $\delta_2 = -\delta_1$ (dashed line). $\rho_c = 1$ corresponds to the critical intensity for the OFT (no thermomechanical effect).

In the calculations, we took the material parameters for the nematic E7: $K_3 = 15.97 \times 10^{-12} \text{ N}$, $n_e = 1.746$, $n_o = 1.522$ (extraordinary and ordinary refractive indices), $\lambda = 514 \text{ nm}$, $\alpha_\perp = 88 \text{ cm}^{-1}$ (absorption coefficient), $\kappa_{||} = 10^{-3} \text{ W/}^\circ\text{C cm}$ (the heat conductivity for the nematic). The calculations were made for a layer of $75 \mu\text{m}$ thickness. For these parameters and for the threshold value $I_F = 32 \text{ W/cm}^2$ observed in our preliminary experiment, ζ turns out to be $\zeta \simeq 52$ which is a typical value for the enhancement factor [17] [see also Eq. (5)]. The maximal temperature difference inside the layer is $\Delta T_{max} \simeq 20 \text{ K}$. In the calculations we used $b = 0.8$ and $\alpha'_2 = -1.058$. The thermomechanical coefficients and, hence, δ_1 and δ_2 are unknown. Thus we took the typical order of their magnitudes as $10^{-12} \text{ N/}^\circ\text{C}$ reported in [6].

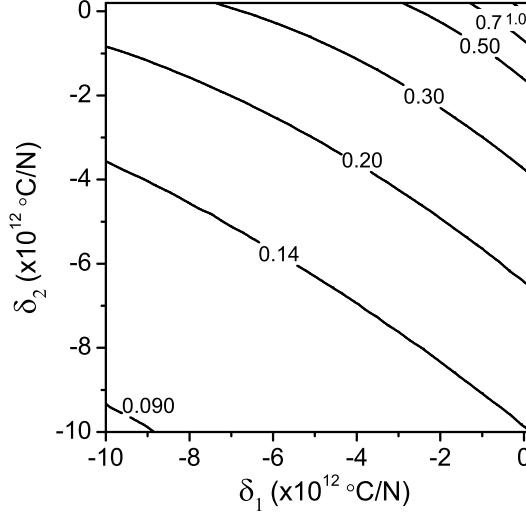


FIG. 3: Contour lines for the surface $\rho_c(\delta_1, \delta_2)$. The values of ρ_c are shown on contour lines. ($\rho_c = 1$ for $\delta_1 = \delta_2 = 0$ corresponds to the case without thermomechanical effect.)

In Fig. 2 the results of numerical solution of Eq. (20) [or alternatively the eigenvalue problem (17)] are shown for three different cases when i) $\delta_2 = \delta_1$; ii) $\delta_2 = 0$; iii) $\delta_2 = -\delta_1$. ($\delta_2 = 0$ corresponds to the thermomechanical single-constant approximation.) One can see from this figure that ρ_c might be indeed several times lower than that for the OFT. This fact is clearly demonstrated in Fig. 3 where the contour lines for the surface $\rho_c(\delta_1, \delta_2)$ are plotted. One is now forced to conclude that the thermomechanical effect leads indeed to the substantial change of the critical intensity.

E. Thermomechanical effect due to temperature difference at the boundaries

We have assumed so far that the temperature on both the bounding plates is the same. As a next step, we have analyzed the influence of the nonzero temperature difference ΔT maintained at the boundaries to the instability threshold. To obtain this effect in pure form we assumed that the thermomechanical effect is due to ΔT only and the absorption inside nematic is neglected. For this simple situation the temperature gradient inside nematic is constant. Following the similar procedure described in previous section when linearizing basic equations around the homeotropic state [see Eq. (17)], the following ODE for n_x has

been derived

$$\begin{aligned} \partial_z^2 n_x + [\rho - \sigma(1 - b)] n_x - d_3 \Delta T \partial_z n_x - \\ \frac{b \sigma}{\pi} \int_0^\pi n_x dz = 0, \end{aligned} \quad (21)$$

where

$$d_3 = \delta_1 \eta, \quad \eta = \frac{\kappa_s L}{(\kappa_s L + 2\kappa_{||} d) \pi K_3}, \quad (22)$$

and the other quantities have the same meaning as before. After substituting $\sigma = 0$ in Eq. (21) the following simple formula for the critical intensity was found

$$\rho_c = 1 + d_3^2 \frac{(\Delta T)^2}{4}. \quad (23)$$

As is seen ρ_c is always higher than the threshold for OFT and depends quadratically on ΔT . This effect is, however, small because the thermomechanical coefficient enters quadratically to the expression for ρ_c as well. (The order of magnitude of d_3 is $10^{-2} \text{ } ^\circ\text{C}^{-1}$ for the parameters used in calculations.)

III. CONCLUSIONS

In conclusion, we have shown theoretically that thermomechanical effects might be at the origin of significant lowering of the OFT threshold expected for dye-doped nematic liquid crystals. To explain this, we have developed a simple model, assuming that all physical quantities depend only on the coordinate across the layer. We linearize both the director and Navier-Stokes equations around the basic state, to assess the change of the primary instability due to thermomechanical effects. The temperature gradient across the layer, which is induced by light itself due to absorption of the dye dopants, was calculated from the 1D heat equation. Using a typical value for the thermomechanical coefficients, we have found that the effect of OFT's lowering might be explained by thermomechanical effects. We have also analyzed a situation when the thermomechanical effects are due to the temperature difference maintained at the boundaries. It turned out that in this case they always lead to an increase of the OFT threshold.

IV. ACKNOWLEDGMENTS

This paper is dedicated to the memory of Professor Lorenz Kramer who suddenly passed away on 05/04/2005.

The authors are grateful to Dr. Gabor Demeter for his helpful discussions. D. K. gratefully acknowledges financial support by the Deutsche Forschungsgemeinschaft under Kr 690/16.

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